



Understanding the Ag/Al₂O₃ hydrocarbon-SCR catalyst deactivation through TG/DT analyses of different configurations

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ABSTRACT

Understanding the parameters affecting the Ag/Al₂O₃ hydrocarbon SCR catalyst activity or the procedure by which the C-containing species are deposited on the catalyst surface, under actual diesel engine operation, can lead to design a low temperature active Ag/Al₂O₃ SCR catalyst. In this work we investigated the role of the Ag/Al₂O₃ HC-SCR catalyst configuration for NO_x reduction activity and its deactivation by coking under passive and active operation. This was done by separating the powdered catalyst into two layers/partitions (front and rear). The single and double-layered catalysts were exposed to real diesel engine exhaust gas, and then thermo gravimetric (TG) and differential thermal (DT) analyses were carried out. The analysis showed that the retention of carbon-rich species (hydrocarbons and soot) in the double-layered configuration occurs mainly in the first catalyst layer (front layer), therefore resulting in a cleaner second layer (rear layer). The results confirm that the catalyst deactivation initiated at the front part of the catalyst and progressively spreads towards the back. TG/DT analyses also showed that the retention of carbon-rich species over the catalysts was strongly reduced at temperatures higher than 350 °C or when hydrogen was introduced, but the deactivation pattern was similar. The reactor design (e.g. catalyst configuration) in HC-SCR with silver is an important factor, and in the presented work the space in between the beds shown to be a critical parameter as it enhanced the low temperature catalytic activity.

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1. Introduction

Selective catalytic reduction of NO_x with hydrocarbons (HC-SCR) has been extensively researched and developed as a potential method for the removal of NO_x under oxygen-rich conditions. Among the many HC-SCR catalysts tested under laboratory and real engine exhaust conditions, silver catalysts are widely accepted as the most promising. This is due to their excellent ability to reduce NO_x with a wide variety of hydrocarbons, such as oxygenated hydrocarbons [1,2], light hydrocarbons [3,4] and heavy hydrocarbons [5,6]. In addition, it has also been reported that the water vapour, present in the diesel exhaust gas, inhibits the unselective oxidation of hydrocarbons, therefore promoting the catalyst NO_x reduction activity [7].

However, high exhaust temperatures are required for good NO_x reduction efficiencies using silver catalysts. As a result, the main

drawback of silver catalysts is their lack of low temperature activity. Although, their activities can be improved at low temperatures when long chain hydrocarbons are used [7,8], this is not enough to achieve acceptable catalyst activity unless hydrogen is present. Hydrogen is widely considered as an ideal co-feeder gas in active mode (i.e. injected HCs) HC-SCR operation in order to decrease the minimum temperature needed to drive the NO_x-reducing reactions [9,10]. However, the hydrogen mechanism for reducing the catalyst activity window to lower temperatures is not fully understood yet. For example, Shibata et al. [11] found that the addition of hydrogen resulted in remarkable promotion of hydrocarbon to mainly surface acetate, which is the rate-determining step of hydrocarbon-SCR in the absence of hydrogen. Later, Shimizu et al. [12], after conducting some kinetic studies, reported that hydrogen addition results in a decreased activation energy for NO_x reduction (i.e. the temperature window was shifted towards lower values). They also showed that the addition of hydrogen can retard poisoning by reducing the concentration of nitrates on the catalyst surface, therefore enhancing the HC-SCR reaction dramatically. On the other hand, Houel et al. [6] suggested that the addition of hydrogen promotes the forma-

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tion of NO_2 over the $\text{Ag}/\text{Al}_2\text{O}_3$ catalyst, which is very effective in oxidising carbon-rich species at low temperature, thus preventing catalyst deactivation.

The effect of $\text{HC}_1:\text{NO}_x$ ratio on the silver catalyst activity has also been studied. Eränen et al. [13] reported that an optimum value of $\text{HC}_1:\text{NO}_x = 6$ is required. Recently, Houel et al. [6,8] found that the selection of an optimum $\text{HC}_1:\text{NO}_x$ ratio depends on the diesel engine exhaust temperature and hydrocarbon speciation. Normally, a suitable $\text{HC}_1:\text{NO}_x$ ratio falls between 1 and 5 in the temperature range of 200–450 °C.

Other researchers have shown that the gas phase reactions occurring downstream of the silver catalyst can play an important role in the overall NO_x reduction activity. For example, Eränen et al. [14] studied these reactions under different configurations and they found that the distance between the silver catalyst and the diesel oxidation catalyst (DOC) (the latter located behind the former) significantly affects the gas phase reactions. The conversion of NO_x to N_2 decreased substantially when the DOC was placed immediately after the silver catalyst, but when the distance was increased, the NO_x conversion increased too. They concluded that the reduction of NO_x over $\text{Ag}/\text{Al}_2\text{O}_3$ does not only take place on the surface of the catalyst but also in the gas phase. Klingstedt et al. [15] also studied the effect of catalyst configuration on NO_x conversion efficiency. In their studies, when the silver/alumina catalyst was divided into four segments, the activity (NO_x reduction) at low temperatures (250–350 °C) was improved compared to that of a single segment. They suggested that the empty space between each segment enabled gas phase reactions to take place and thus, higher NO_x conversion efficiencies were achieved. Although, it is well known that the deposition of carbon-rich species on the catalyst surface is a major problem associated with low temperature HC-SCR; activity improvements observed in layered configurations [e.g. 15] could also be due to a different accumulation rate of poisonous species

on each segment, thus slowing down catalyst deactivation of rear layers.

In our earlier work [6], we reported that if a single-component hydrocarbon (n-octane, n-decane and n-dodecane) is used as a reducing agent there is no evidence of catalyst deactivation with/without hydrogen. However, when long chain multi-component fuels (i.e. conventional and synthetic diesel) are used, the presence of hydrogen does not prevent C-containing species accumulation at high $\text{HC}_1:\text{NO}_x$ ratios. In the present work, powdered $\text{Ag}/\text{Al}_2\text{O}_3$ catalyst activity and deactivation over time were examined under both passive (no fuel injection) and active (with fuel injection) operation coupled with/without hydrogen addition. We examined the effect that the catalyst configuration (placed in a single or double layer) may have on the deactivation process using thermogravimetric analysis (TGA) of the different catalyst layers. It provided clear evidence of a different deposition rate of carbon-rich species, with a different composition (soot-to-hydrocarbon ratio) depending on the catalyst configuration.

2. Experimental

A silver catalyst (2 wt%) was prepared by impregnating γ -alumina with an aqueous solution of silver nitrate (AgNO_3), before drying (16 h) and calcining (in air for 2 h at 500 °C). It was prepared as a powder, which was then granulated to a particle size of 250–355 μm for packed-bed testing. For a single-layered catalyst configuration, a 0.6 g bed of the catalyst was filled into a mini-reactor. For the double-layered configuration, the same mass of catalyst was separated into two layers of 0.3 g each. A simplified schematic of the catalyst reactor system used is shown in Fig. 1. The flow rate at the pump was adjusted to fix a space velocity in the catalyst of 30,000 h^{-1} . The SCR mini-reactor was heated by a tubular furnace and its temperature was set by a thermocouple

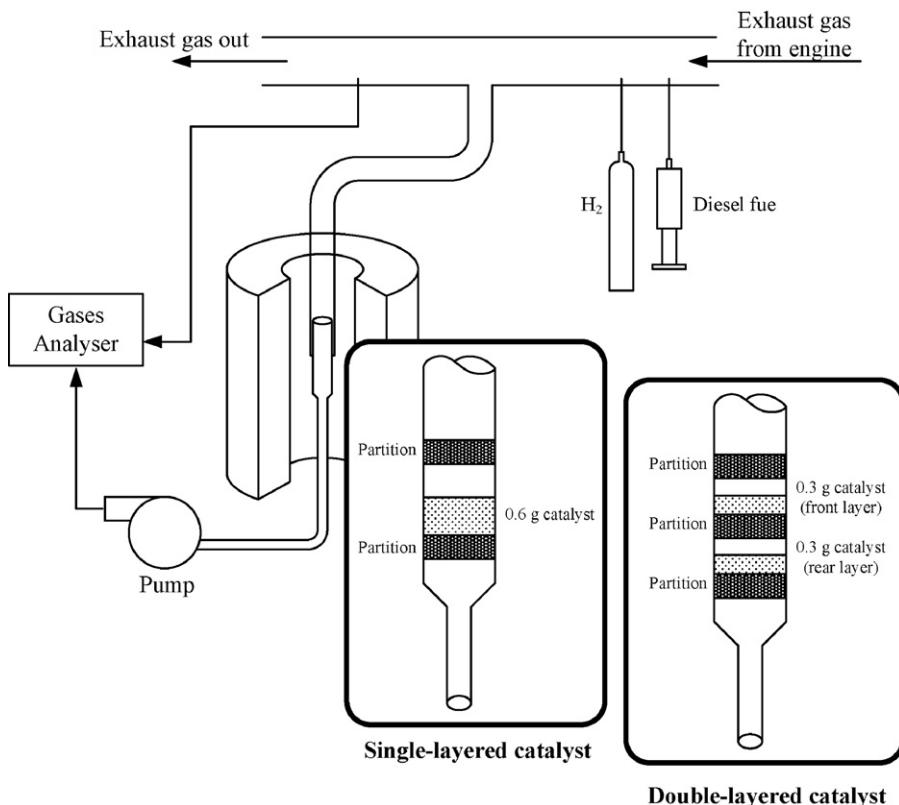


Fig. 1. Simplified schematic of the catalyst development rig.

Table 1
Fuel properties.

	Method	ULSD fuel
Cetane number	ASTM D613	53.9
Density at 15 °C (kg/m ³)	ASTM D4052	827.1
Viscosity at 40 °C (cSt)	ASTM D 445	2.467
50% distillation (°C)	ASTM D86	264
90% distillation (°C)	ASTM D86	329
LHV (MJ/kg)		42.7
Sulphur (mg/kg)		46
Aromatics (wt%)	ASTM D2622	24.4
C (wt%)		86.5
H (wt%)		13.5
H/C ratio		1.88

positioned at the inlet of the reactor, 5 mm upstream of the catalyst bed.

The exhaust gas was generated by a single-cylinder, direct injection diesel engine (a Lister Petter TR1 engine). The key specifications of this engine have been presented in earlier work [16,17]. The engine was run on ultra-low sulphur diesel (ULSD) fuel, whose main physical and chemical properties are listed in Table 1. An AVL Digas4000 analyser was used to measure the emissions from the exhaust gas, including O₂ by electrochemical method, CO and CO₂ by non-dispersive infrared (NDIR). Emissions of NO_x were measured by chemiluminescence and total hydrocarbons (HC₁) using a heated flame ionization detector (FID). All analyzers were properly calibrated with certified bottled gases.

Some of the tests were performed with hydrogen addition from a certified bottled source with a high purity (>99%). Under active SCR operation diesel fuel was injected in the exhaust gas using a medical syringe pump and a fuel injection system that comprised a fuel atomizer.

A Perkin Elmer Pyris Diamond TG/DTA Thermogravimetric/Differential Thermal Analysis Module was used. The temperature range with the used module is from ambient temperature to 1100 °C. The temperature program consisted in an initial heating step in N₂ atmosphere up to 400 °C (5 °C ramp rate), then the gas was switched to air (oxidant atmosphere) keeping constant the ramp rate at 5 °C/min up to 630 °C. Finally the sample was held at 630 °C for 10 min. Following the manufacturer instructions, the DTA module was calibrated in advance to the start of all the tests in order to find the conversion between the temperature difference given by the DTA and the heat rate from/to the sample (expressed in mW), which is plotted in the graphs presented in this work.

3. Test procedure

The engine was run in a single steady state operating mode (engine speed: 1500 rpm; indicated mean effective pressure—IMEP: 2 bar). Under this condition, the exhaust gas composition (dry basis) was as follows: 2.7% CO₂, 16.86% O₂, 254 ppm NO_x, 160 ppm HC₁, 100 ppm CO. The H₂O content was estimated to be around 3% when the values were converted to wet basis. For all the tests, 4 l/min of exhaust gas was passed through the mini-reactor using a vacuum pump (see Fig. 1). The furnace temperature was adjusted in order to raise the SCR catalyst inlet temperature from 350 to 450 °C in 50 °C steps.

At each temperature, tests were performed under passive SCR (i.e. no fuel injection in the exhaust gas) and active SCR operation (i.e. with HC injection into the exhaust). Under passive operation the resulting HC₁:NO_x ratio was 0.63 calculated from the exhaust gas composition. Conversely, under active SCR operation two fuel flow rates were examined, namely 15 and 30 ml/h, resulting in an increase of the HC₁:NO_x ratio to 1.38 and 2.13, respectively. These HC₁:NO_x ratios match well with the aforementioned optimized

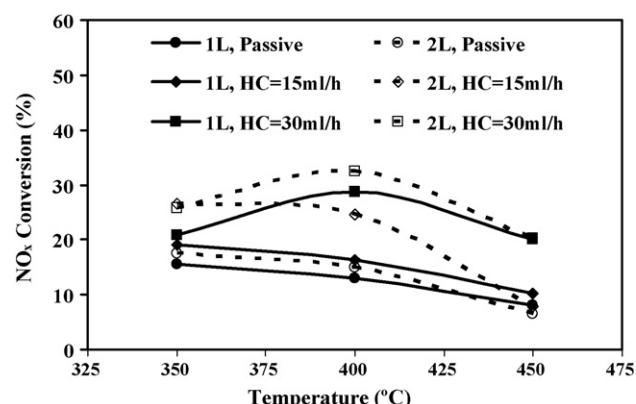


Fig. 2. Effects of hydrocarbon injection (15 and 30 ml/h) on NO_x activity on single (1L) and double-layered (2L) catalyst configuration.

range of between 1 and 5, suggested by Houel et al. [6,8]. The hydrogen effect on SCR performance was evaluated by conducting the tests firstly, with no hydrogen addition (although approximately 120 ppm hydrogen was produced from the engine combustion process) and secondly, by adding 1500 ppm of hydrogen (concentration measured in the exhaust gas after adding hydrogen from the bottle). During each test, NO_x measurements were taken for 10 min from the locations indicated in Fig. 1, at 1 min intervals, in order to evaluate the activity/deactivation of the catalyst over that particular space of time.

After carrying out the tests, catalyst samples of 6 mg were loaded into a Pyris TG/DTA analyser and evaluated in order to determine the amount of soot and hydrocarbons deposited on the catalyst surface.

4. Results and discussion

4.1. Effects of temperature, fuel injection and catalyst configuration

Fig. 2 compares the Ag/Al₂O₃ SCR catalyst activity in reducing NO_x emissions under passive and active (fuel injection) operation, for the single and double-layered catalyst configurations. The activity values plotted in this figure correspond to the calculated activity after 10 min of exhaust flow through the catalyst.

Regarding the effect of the catalyst configuration, in most cases, the double-layered configuration resulted in increased NO_x conversion (Fig. 2) activity. This, as suggested by Klingschedt et al. [15], may be due to the formation of nitrogen not only on the double-layered catalyst surface but also in the gas phase in the empty space between the front and the rear layers. In addition, the double-layered catalyst improves the diffusion of the SCR reactants onto the active sites of the catalyst. As it is explained later in the paper, a different accumulation rate of poisoning species in the front and rear layers occurs, but this is likely to happen in the single-layer configuration as well. For the single layer, the poisoning species are deposited in the front and then spread slowly towards the back of the catalyst. Hernández Carucci et al. [18] using diesel type fuel reported that the condensation of hydrocarbons on the catalyst surface ceases the oxidation of hydrocarbons, which is a key step on the SCR reactions. With our results we can conclude that both effects are initiated at the front of the packed powder catalyst and progressively spreads towards the back. In addition, other species formed on the catalyst may contribute in the two above described effects.

Interestingly, the aforementioned effect of the catalyst configuration was not uniform across the whole temperature range. The benefits of the double-layered configuration declined with the

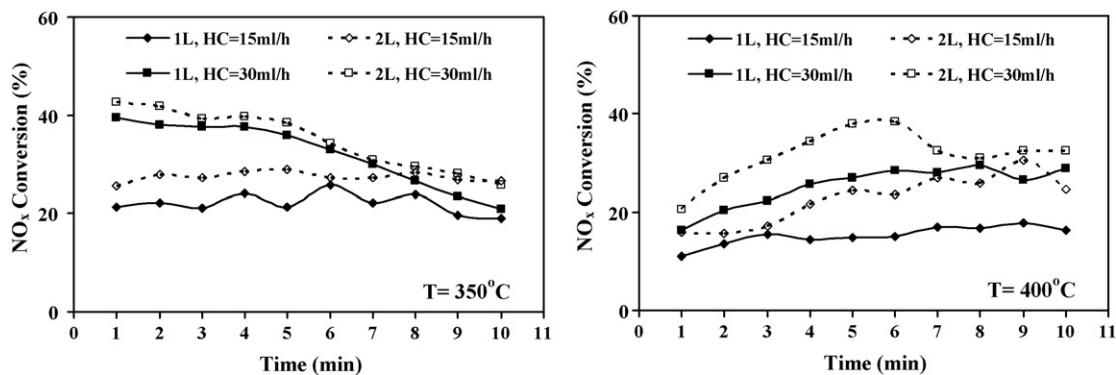


Fig. 3. Effects of hydrocarbon injection (15 and 30 ml/h) on NO_x activity on single and double-layered catalyst configuration at isothermal conditions.

increasing temperature (Fig. 2), with no benefits at the temperature of 450°C . This is because at this temperature there is minimum deposition of poisoning species on the catalyst surface regardless of its configuration.

Diesel fuel injection improved the catalyst activity regardless of the catalyst operating temperature, although this improvement was more evident at 400°C . Increasing temperature from 400 to 450°C resulted in a decrease in the NO_x conversion under all the conditions tested. This indicates that at 450°C hydrocarbons are predominantly selected for oxidation reactions rather than NO_x reduction reactions. However, when the temperature was increased from 350 to 400°C , the NO_x conversion trend, showed more dependence on the amount of fuel injected. When no fuel or the lower amount of fuel (flow of 15 ml/h) was used, the NO_x conversion decreased with temperature (it is assumed that the hydrocarbons present were either oxidised or deposited on the catalyst surface). But with the 30 ml/h fuel flow, enough fuel is present to promote NO_x reduction over the catalyst.

Fig. 3 shows the trend of the catalyst activity over time at isothermal conditions. It is clear from this figure that the double-layered configuration showed a higher activity. At a relatively low exhaust temperature (350°C), the catalyst activity decreased over time when a 30 ml/h fuel was injected in both single and double-layered catalyst configurations. The deactivation of the catalyst did not occur when the amount of fuel was reduced and/or the temperature was increased (Fig. 3). Most likely, at 350°C the 30 ml/h fuel injection exceeded the optimum quantity, resulting in a higher deposition rate of hydrocarbons onto the catalyst surface. To validate this assumption, a thermogravimetric analysis (TGA) of the single and double-layered catalysts was performed at temperature of 350°C , for 15 ml/h and 30 ml/h fuel injection (Fig. 4). This analysis was used to find the quantity of the carbon-rich species, i.e. volatile organic compounds (VOCs) and soot that were deposited on the silver catalyst surface.

In the graphs presented in Fig. 4 (and all the analogous figures in the paper), we assumed that at the TGA-temperature range of

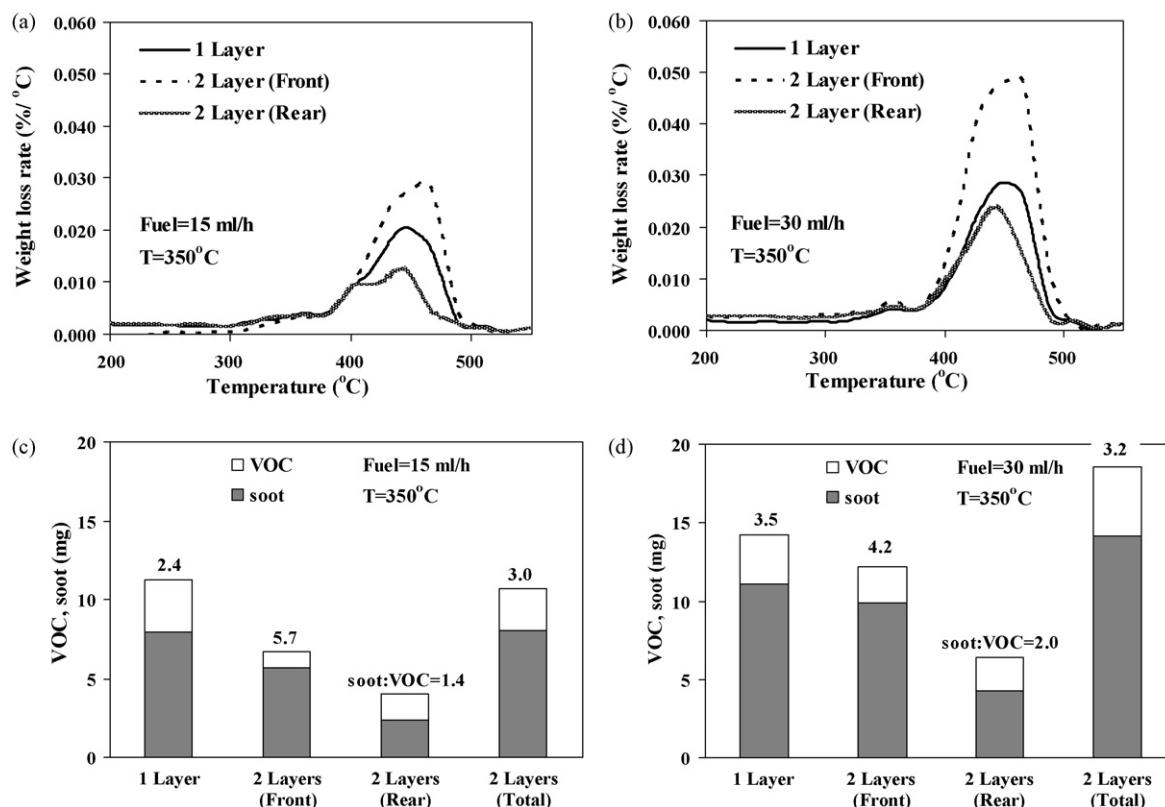


Fig. 4. TG analysis of the single and double-layered (front and rear) catalyst at 350°C for 15 ml/h (a and c) and 30 ml/h (b and d) hydrocarbon injections.

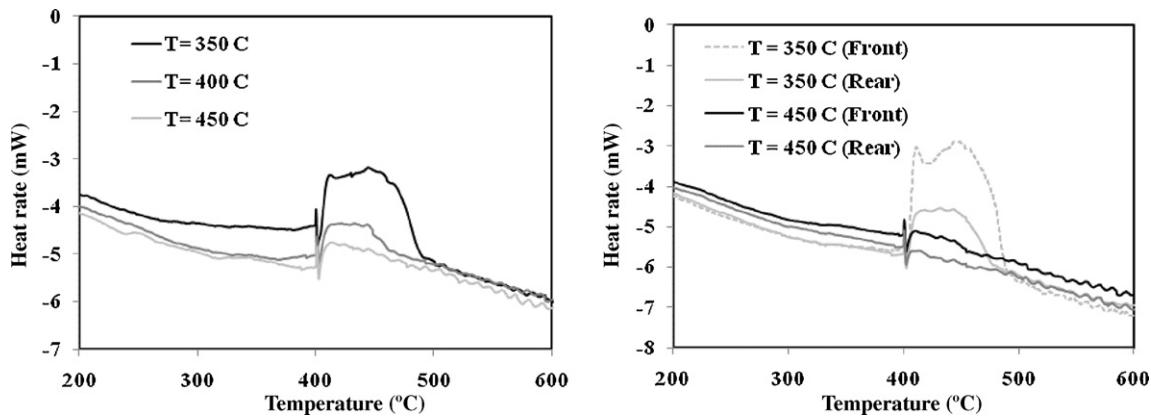


Fig. 5. DT analysis effects of exhaust temperature on carbon oxidation over a single (left) and double (right) layered catalyst, fuel injection 30 ml/h.

200–400 °C (using inert atmosphere, as explained in Section 2) the weight loss corresponds mainly to the vaporization of the VOC that are deposited on the catalyst. Then, between 400 and 630 °C (using an oxidising atmosphere) the weight loss represents the amount of soot accumulated on the catalyst surface. We decided not to consider the weight loss from the initial temperature (~30 °C) up to 200 °C, as it was mostly due to the water evaporation and it can be affected by the initial transient period and the pan movement inside the TGA furnace, resulting in non-accurate data [19]. In Fig. 4c and d, the mass values represented were calculated from the TGA weight loss rate and the total catalyst mass in the different catalyst configurations and layers. In addition, the soot-to-VOC ratio is also presented in the graphs.

It is shown in Fig. 4a and b that the weight loss mostly occurs in the TGA-temperature range of 400–500 °C. This indicates that

soot is the main compound deposited on the catalyst surface. As expected from the earlier discussion, the total mass of carbon-rich species (VOC and soot) increased as the fuel flow was increased, see Fig. 4c and d. This justifies the rate of catalyst deactivation discussed in Fig. 3. When only 15 ml/h of fuel was injected there was not enough hydrocarbons to promote NO_x conversion (Figs. 2 and 3), but at the same time the low fuel flow rate prevented catalyst deactivation (Fig. 3), resulting in a reduced amount of adsorbed species on the catalyst surface (Fig. 4).

The higher deposition of soot on the front layer (Fig. 4) reflects that (i) the front layer acts as a soot/particulate matter filter, and consequently it prevents the rear layer from deactivation, and (ii) the catalyst deactivation initiates at the front and progressively spreads towards the back of the catalyst bed, (iii) the reactive intermediates generated in the front layer promote the gas phase

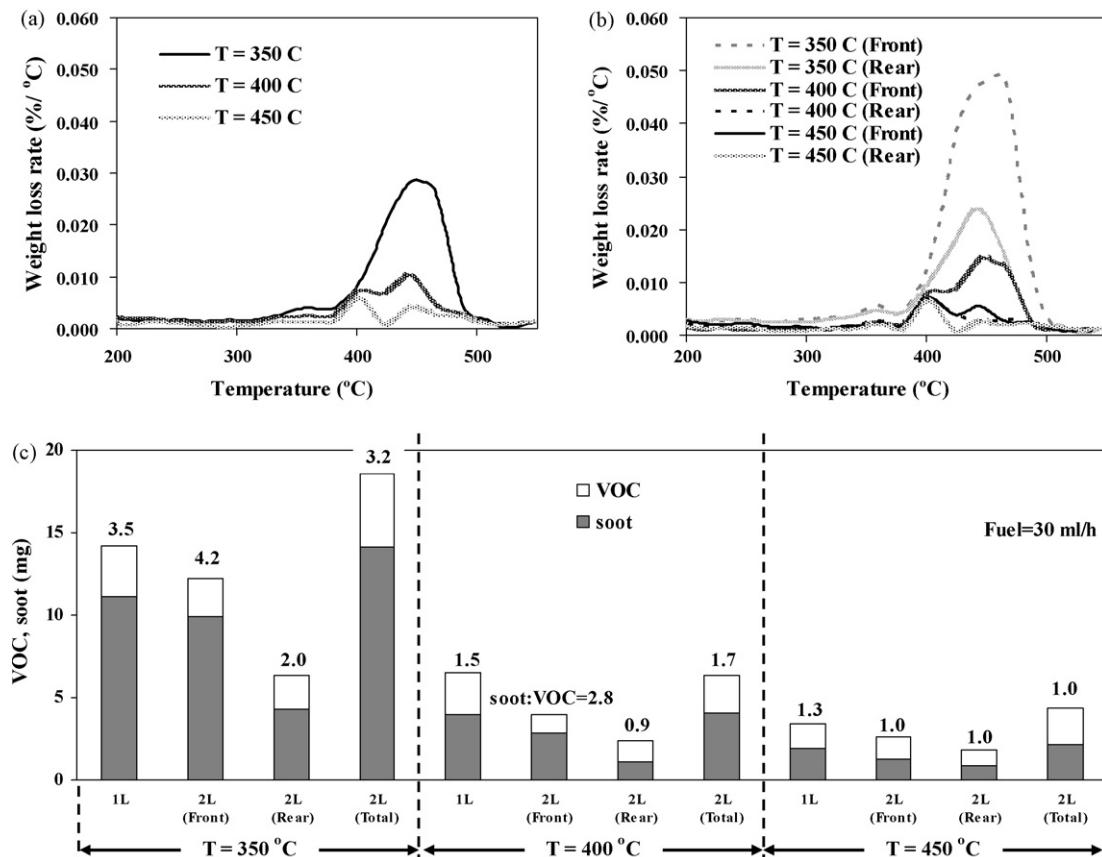


Fig. 6. TG analysis effects of exhaust temperature on carbon oxidation over a single (Fig. 5a) and double (Fig. 5b) layered catalyst, fuel injection 30 ml/h.

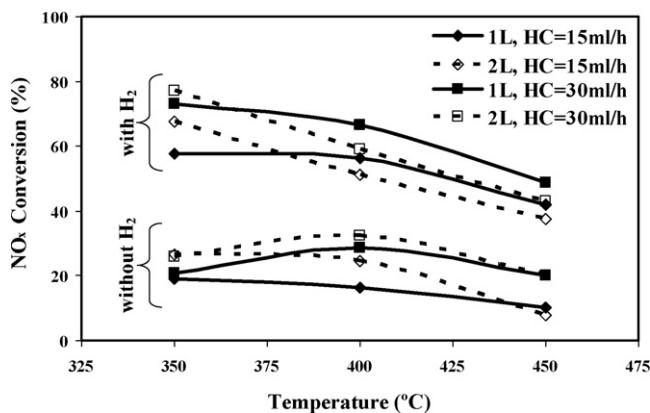


Fig. 7. Effects of hydrocarbon injection (15 and 30 ml/h) on NO_x activity on single and double-layered catalyst configuration, hydrogen 1500 ppm.

reactions in the empty space between the front and rear layers. Since a higher mass of compounds is deposited in the front layer compared to the rear one (double layer configuration), it can be assumed that in the single-layer configuration the compounds are accumulated mainly at the front, preventing the rest of the catalyst from being poisoned. Accepting that this “filtering effect” happens regardless of the catalyst configuration, the higher NO_x conversion showed by the double layer configuration has to be due to the gas phase reaction taking place in the empty space between layers, as suggested by Klingstedt et al. [15] and previously discussed. To further support this argument, it can be seen in Fig. 4c and d that the total mass of VOC and soot deposited on the catalysts is approximately the same or even higher in the double-layered configuration (fourth bar of Fig. 4 c and d) than in the single-layered one (first bar). Nevertheless, the conversion of NO_x over the double-layered catalyst was higher, as shown in Figs. 2 and 3.

The specific effect of the test temperatures is depicted in Figs. 5 and 6. As expected, the weight loss dropped sharply when the temperature was increased. At the lowest test temperature (350 °C), the sample weight loss was the highest independently of the layer configuration (Fig. 6), i.e. single or double, being this effect more notorious when the TG/DTA atmosphere was switched to oxidant. This weight loss in the 350 °C test occurred together with the highest recuperation in the temperature difference between the reference material and the sample (see DTA plots in Fig. 5), meaning that the sample weight loss under oxidant atmosphere is mainly due to oxidation of deposited compounds, which is an exothermic process, the heat release suggests the reduction of these compounds calculated from the temperature difference between the reference and sample point. In both catalyst configurations, soot

and VOC depositions were virtually eliminated when the temperature was 450 °C. Fig. 6c also shows two interesting trends with regard to the soot-to-VOC ratio. Firstly, the ratio was reduced by increasing the temperature due to the soot being oxidised when sufficient activation energy (i.e. high temperatures) was provided. Secondly, the soot-to-VOC ratio in the front layer was higher than that of the second layer, which is explained by the filter-like behaviour, commented on earlier, for the front layer. Unfortunately, although the soot-derived poisoning is sharply reduced at higher temperature, NO_x conversion is also reduced (Fig. 2) due to more hydrocarbons being selected for the oxidation reaction rather than NO_x reduction. To maintain or further improve NO_x conversion, the injection of hydrocarbons would need to be increased too.

4.2. Effects of hydrogen

The conversion rates of NO_x over the Ag/Al₂O₃ catalyst, in the temperature range of 350–450 °C, when 1500 ppm hydrogen was added to the exhaust gas are presented in Fig. 7. NO_x emissions conversion was significantly increased when hydrogen was injected. On average, NO_x conversion with hydrogen approximately doubled the conversion without hydrogen, and in some of the conditions 80% reduction was observed.

It can also be observed from Fig. 7 that, in all the tests with hydrogen addition, the NO_x conversion was decreased with the increasing temperatures. It is known that the addition of hydrogen chemically promotes both hydrocarbon oxidation and NO_x reduction in the HC-SCR mechanism. It seems that, when the temperature was increased to 450 °C, the injected hydrocarbons were mainly consumed by the oxidation reaction rather than promote the NO_x reduction reaction. In conclusion, the optimal hydrocarbon-to-NO_x ratio appears to be a function of the catalyst temperature (as proposed by Houel et al. [6,8]) as well as the presence of hydrogen both of these factors should be optimized together.

Interestingly, at 350 °C the double-layered catalyst provided a considerably higher NO_x conversion (increase of 5–10%) compared to that over the single-layered configuration. However, the trend was reversed as the temperature increased, because higher temperatures resulted in cleaner front and rear layers (for the double-layered configuration) that promoted the oxidation of hydrocarbons in the presence of hydrogen, as it has been previously reported [21].

By comparing Figs. 3 and 8 it is clear that hydrogen can eliminate, or at least strongly reduce catalyst deactivation under the most adverse conditions (high fuel flows and low temperatures). To further prove this point, Fig. 9 shows the TG analysis of the single and double-layered configuration at 350 °C and with 1500 ppm hydrogen addition. The trends are roughly the same as those described

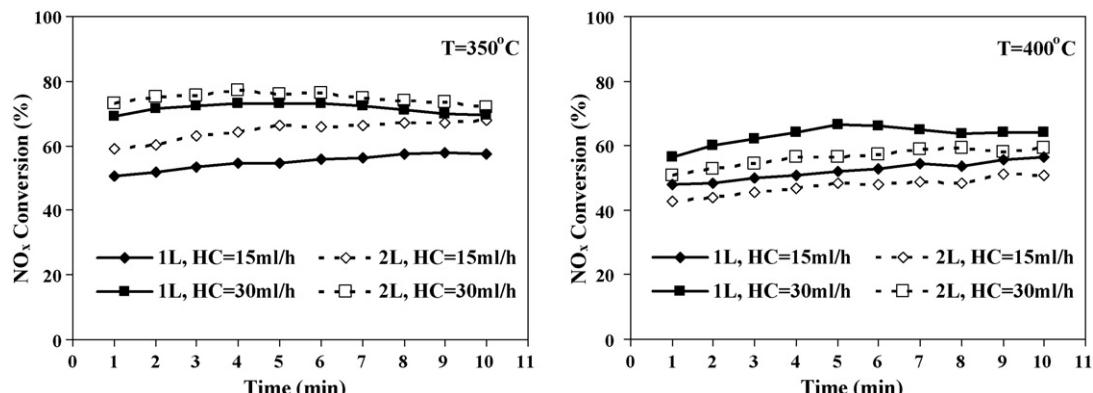


Fig. 8. Effects of hydrocarbon injection (15 and 30 ml/h) on NO_x activity on single and double-layered catalyst configuration under isothermal conditions.

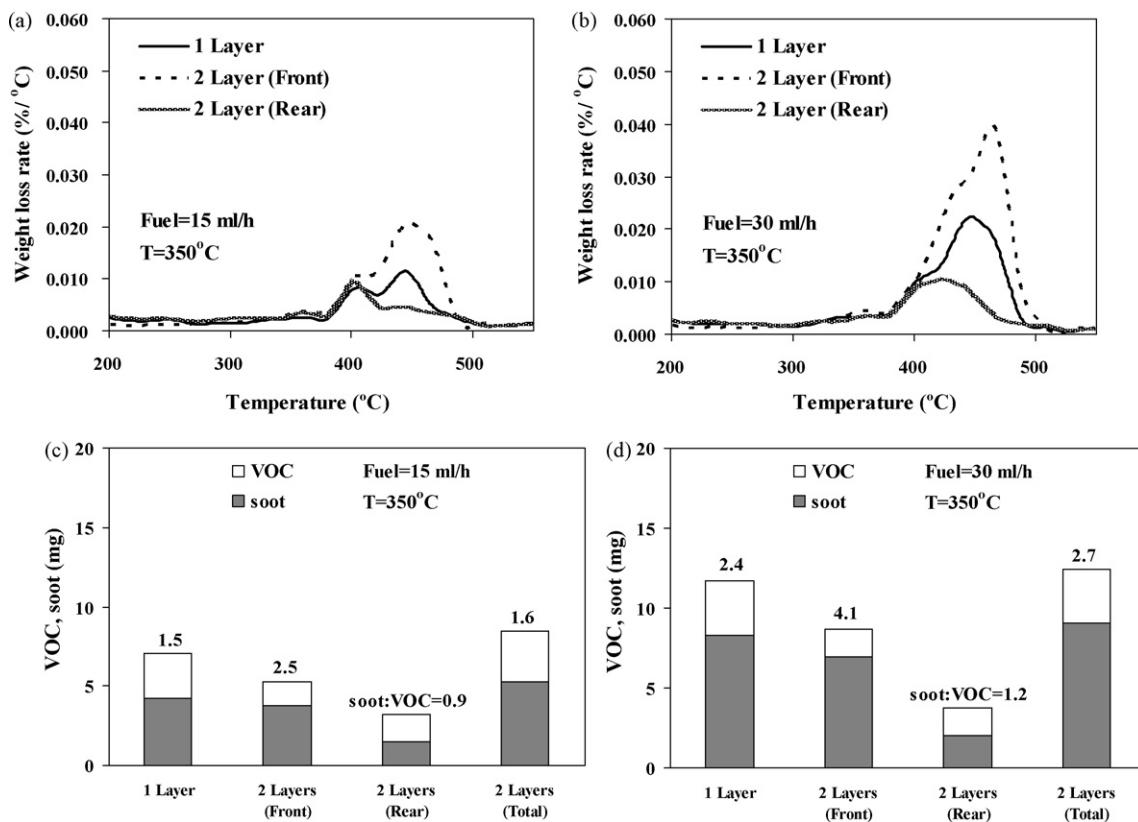


Fig. 9. TG analysis of the single and double-layered (front and rear) catalyst at 350 °C for 15 ml/h (Fig. 8a and c) and 30 ml/h (Fig. 8b and d) hydrocarbon injections, hydrogen 1500 ppm.

for the tests without hydrogen in Fig. 4 (soot being the main compound deposited on the catalyst surface), but the mass of carbon-rich species is sharply reduced with the hydrogen addition. This explains why the NO_x conversion did not fall over time with 30 ml/h fuel flow. It is obvious from these graphs that hydrogen significantly reduces the amount of C-containing species adsorbed on the catalyst surface, and this may occur by enhancing hydrocarbons oxidation and hence reducing the rate of fuel condensation on the catalyst surface. However, for the hydrogen additions tested, hydrogen does not affect the pattern of the deactivation (i.e. starting at the front and moving towards the back).

Furthermore, the reduction in the carbon-rich species deposited onto the catalyst surface was seen in all the conditions tested, as summarized in Fig. 10. This reflects the fact that hydrogen pro-

motes the oxidation of the species collected on the catalyst surface in both single and double-layered configurations, thus increasing the number of active sites available to catalytically reduce NO_x and consequently increase NO_x reduction activity (Fig. 7). It is worth mentioning that the addition of hydrogen also decreased the soot-to-VOC ratio. This trend points out the fact that, with regard to the oxidation reactions, soot oxidation is more strongly promoted over the catalyst than hydrocarbon oxidation, possibly through the NO₂–soot oxidation [20]. We have previously reported [21] that hydrogen addition promotes NO_x oxidation to NO₂ over Ag/Al₂O₃-SCR in a similar temperature range to the one examined in this study. As a result, more hydrocarbons are available to further increase the NO_x reduction activity through HC-SCR reactions.

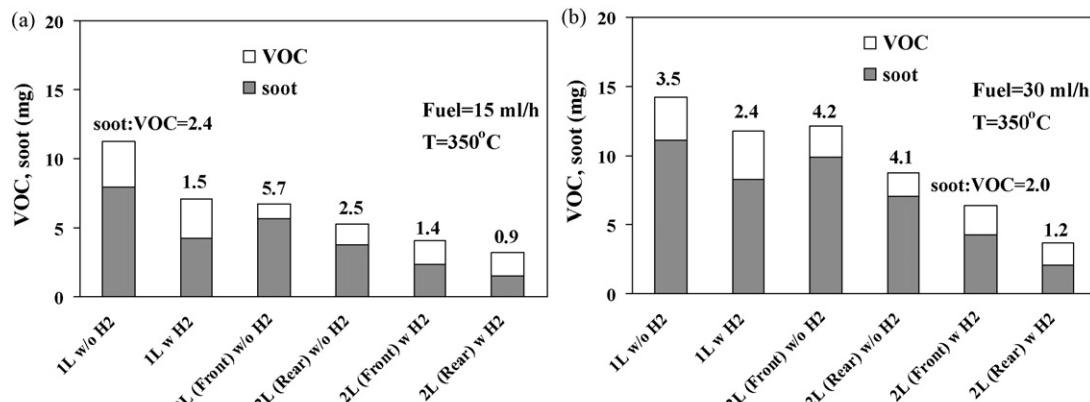


Fig. 10. Comparison the effects of hydrogen addition to soot and VOC at 350 °C for 15 ml/h (Fig. 9a) and 30 ml/h (Fig. 9b) hydrocarbon injections.

5. Conclusions

The Ag/Al₂O₃ HC-SCR catalyst deactivation and NO_x conversion were evaluated for different configurations in this work. The findings presented highlight the importance of a proper selection of the SCR catalyst design and the SCR integration with other vehicle aftertreatment systems in order to maximize NO_x reduction activity. In the absence of hydrogen the double-layered configuration showed higher NO_x conversion compared to the single-layer. This result was explained based on the gas phase reaction occurring in the empty space between layers, since the filtering effect of the catalyst is similar in both configurations (single and double layers). The NO_x-reducing reactions probably occur on the catalyst surface but also in the empty space between layers, thus enhancing the NO_x conversion through an optimised disposition of the catalyst mass in different layers. Evidence from TG/DT analyses proved that soot is the main component of these carbon-rich species, but hydrocarbons are also deposited. The front bed of the double-layered catalyst prevented the rear bed from accumulating excessive hydrocarbons and soot; subsequently, the second layer remained cleaner and more active sites were available for NO_x to N₂ reduction. In vehicle applications this effect may not be reproduced by using two SCR monoliths to mimic the two layers, but it can be reproduced by positioning the SCR catalysts downstream of a diesel particulate trap or diesel oxidation catalyst where large amounts of the engine soot and hydrocarbons will be reduced before they reach the SCR catalyst. The need of hydrocarbons in the SCR catalyst can be achieved by fuel injection upstream the catalyst or by the addition of hydrogen-rich gas that contains a range of hydrocarbon species (e.g. diesel fuel reformate). As our results revealed, when hydrogen was added, the optimum catalyst configuration was dependant on the operating temperature. At higher temperatures, the double-layered configuration yielded lower NO_x conversion compared to that of the single-layered catalyst because the hydrogen addition promoted the oxidation of hydrocarbons over the cleaner front and rear catalyst layers. Under active SCR system operation, parameters such as hydrocarbon-to-NO_x and hydrogen-to-hydrocarbon ratios should be selected based on the exhaust gas temperature.

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